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Line Shapes in Laser Experiments.

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Abstract

A brief review of recent developments in line shape studies using laser spectroscopy is presented. Experiments are discussed in which the absorption or emission line profiles associated with various transitions in a vapor can be used to provide useful information concerning the atomic sample being tested. In particular, ways in which collisional effects manifest themselves in line shapes are described and the manner in which collision cross section can be extracted from these line shapes is discussed. Illustrative experiments using methods of linear spectroscopy, two-photon spectroscopy and saturation spectroscopy are presented.

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# 1. Introduction

With the rapid development of tunable lasers, there has been a renewed interest in the field of atomic spectroscopy. Not only has traditional spectroscopy been carried out with increased precision using laser sources, but also new spectroscopic techniques for probing atomic systems have been developed. Historically, one has studied absorption or emission line shapes to determine atomic transition frequencies, lifetimes, and oscillator strengths. Moreover, line shape studies have been used to provide information pertaining to collisional processes occurring in vapors. With the laser revolution, one can now expect to obtain new and valuable data concerning both atomic structure and collisional effects by studying laser spectroscopic line shapes.

In this brief review it is not possible to discuss all the recent developments in laser spectroscopy. Therefore, we limit the discussion to a few cases of current interest. In particular we emphasize the experimental aspects of linear and saturation spectroscopy using weak laser fields (Rabi frequency much less than homogeneous widths) in which collisional processes are probed. We assume that collisions can be described by relatively simple models. The collisions are taken to be thermal and describable by the impact theory of pressure broadening -- we consider only binary collisions between atoms and detunings between laser and atomic transition frequencies which are small compared with the inverse duration of a collision.

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Several reviews of collisional studies using laser spectroscopy have recently appeared<sup>1-6</sup> but these reviews did not emphasize the experimental aspects of the problem as we shall do in this work. Section II is devoted to a study of linear spectroscopy, Section III to two-photon spectroscopy, Section IV to saturated absorption and Section V to saturation spectroscopy of three-level systems.

## II Linear Absorption

Linear spectroscopy is a classic technique for studying atomic systems that can be easily carried out using laser sources. One monitors the absorption of a given transition as a function of the frequency of the applied laser field. The laser field is assumed to be weak enough so as not to significantly alter atomic state populations. Profiles in linear absorption are characterized by the following features.

1. Profile intensities are proportional to the difference of populations in the two levels involved in the transition.
2. The line shape is given by the convolution of a Doppler-shifted Lorentzian representing the absorption profile of a given longitudinal velocity sub-class of atoms with the velocity distribution of these atoms.
3. With laser sources, the apparatus function is negligible compared with other sources of broadening.
4. High signal-to-noise ratios are easily obtained.

Collisions within the vapor add new features to the line shapes. The most common effect of collisions is to introduce a pressure and speed-dependent width and shift into the Lorentzian component of the profile. The width can be related to the total cross-section for scattering of the active atoms in the states of the transition by ground state perturbers. It is also possible in certain cases to observe a narrowing of the total profile (Dicke narrowing) with increasing perturber pressure, provided that the collisional interaction for the two states of the transition is nearly identical.<sup>2</sup> We do not consider this narrowing effect in this review since it rarely occurs for electronic transitions. We therefore take line shapes of the form

$$\int \frac{Y(\vec{v})}{Y^2(\vec{v}) + (\Delta + S(\vec{v}) - \vec{k} \cdot \vec{v})^2} G(\vec{v}) d\vec{v} \quad (1)$$

where  $2Y(\vec{v})$  is the speed-dependent homogeneous width ( $\Gamma_{\text{DHN}}$ , natural plus collisional width),  $S(\vec{v})$  is the speed-dependent collisional shift,  $\Delta$  the detuning of the laser frequency from the absorption transition frequency,  $\vec{k}$  the laser propagation vector and  $G(\vec{v})$  the velocity distribution of the active atoms.

If the speed dependence of  $Y$  and  $S$  is neglected, and if  $G(\vec{v})$  is Gaussian, the resultant line shape is termed a Voigt profile.

As an example of linear spectroscopy with lasers, we describe an absorption experiment on the 3.51  $\mu\text{m}$  line of  $\text{XeI}$ .<sup>7</sup> A xenon laser oscillating at 3.51  $\mu\text{m}$  and finely tunable across its gain curve provides a weak laser beam which propagates inside a discharge tube filled with

13<sup>10</sup>. The absorbed or amplified part of the laser flux is recorded as its frequency is scanned. Background signal is eliminated by using a square wave excitation of the discharge and phase sensitive detection. The line profile is analyzed at several pressures according to equation (1) where  $G(\nu)$  is assumed Gaussian and the speed-dependence of  $\gamma$  and  $S$  is neglected. The shift is determined by the position of the symmetry axis of the line profile relative to that of the same transition measured using a reference tube. One fits the profile of the lineshape with two free parameters, the homogeneous and Doppler widths. Although the deconvolution procedure is not unique, consistent results are obtained by assuming profiles of the form (1). The width of the Lorentzian component is shown in Fig. 1 as a function of pressure of either the He, Ne or Ar perturbers. The width of the Gaussian remains constant with pressure, slightly above the temperature of the bath. The different widths plotted in Fig. 1 are linear functions of pressure and converge to the value 4.7 MHz as the pressure approaches zero, in agreement with the expected value of 4.6 MHz. Data taken from reference 7 indicate that shifts are also linear functions of pressure and converge to zero as the pressure approaches zero.

The situation is somewhat more complicated with Xe atoms as perturbers. Fig. 2 shows the width of the Voigt profile, of the Gaussian component, and of the Lorentzian one obtained after profile analysis. The data clearly show that the Gaussian width no longer remains constant with pressure and that there is a corresponding effect on the width of the Voigt profile. However, the width of the Lorentzian remains

a linear function of pressure within experimental accuracy. The cause of the deviation of the Gaussian may be due to a non-resonant transfer of excitation.<sup>7</sup> With Kr atoms as perturbers, a non-resonant excitation transfer leads to severely distorted profiles.<sup>9</sup> Similar experiments have been performed with He and Ne as active atoms.<sup>10,11</sup>

The creation of non-Maxwellian initial state velocity distributions occurs in certain of the above discharge tube experiments and can be strictly avoided only in absorption experiments on transitions originating from the ground atomic state. Although the existence of non-Maxwellian distributions complicates the analysis, there are certain transitions in the rare gases that can be readily studied spectroscopically only by using discharges. It may be noted that one can readily vary the temperature of discharges to study the temperature dependence of collision rates and shifts.<sup>12</sup> The data shown in Fig. 1 and Fig. 2 are restricted to the core of the line (frequency range on the order of several Doppler widths). In this range one is sensitive mainly to collisions with large impact parameters, typically greater than 5Å. In order to probe the short range part of the interatomic potential one must study the far wings of the line.<sup>13,14</sup>

### III. Two-photon Absorption

In linear spectroscopy one is always faced with the problem of deconvoluting a profile into its components. A great emphasis over the past several years has been placed in obtaining spectroscopic techniques

which are essentially Doppler-free; that is, profiles in which the Doppler component is suppressed. One such method that has been developed is that of two-photon Doppler-free spectroscopy.<sup>15-19</sup> The principle involved is easily understood if one writes the condition for absorption of two

photons in the atomic rest frame. If the separation of the two levels of the same parity is  $2h\nu$  and the excitation photons of frequency  $\Omega$  are provided by copropagating ( $c = 1$ ) or counterpropagating ( $c = -1$ ) laser beams, the resonance condition is:  $\Omega(1 - v/c) + \Omega(1 - cv/c) = 2\nu$ . One sees immediately that for  $c = -1$  the Doppler shifts cancel. As  $\Omega$  is varied, an absorption profile characterized by the natural width of the initial and final states of the transition is obtained. In this method each atom contributes to the signal regardless of its velocity. The Doppler-free nature of this signal is provided by a cancellation of Doppler shifts. The profile also may contain a broad background signal arising from absorption of two copropagating photons from one of the laser beams. It can be eliminated by suitable polarization of the beams. It should be noted that by "two-photon" spectroscopy, we are referring to the absorption of two photons of the same frequency between two levels of the same parity with a non-resonant intermediate level.

In the absence of collisions, the two photon absorption operator determines the line shape. It has been shown that this operator is the sum of a scalar operator and a quadrupolar one, leading to two contributions to the line shape. However, in most cases of interest, only one operator couples the two levels and the line shape reduces to a single Lorentzian<sup>20</sup> having the homogeneous width of the two transition

levels. In principle, it is possible to obtain extremely narrow lines, especially if the initial and final states are metastable. There have been many high resolution experiments carried out using this technique.<sup>21</sup>

As in the case of linear spectroscopy, collisions can be incorporated by using a speed-dependent width and shift in the line profile. The line is no longer a true Lorentzian and can be asymmetric. If one neglects the speed dependence of width and shift, one recovers a Lorentzian line shape that is collision broadened and shifted.

Experiments specifically designed to study collisional effects have been carried out initially by Cagnac's group<sup>22</sup>, and subsequently by other groups (References 23, 24). In particular, in Reference 22, one is able to find the variation of the width and shift of two photon excitations of He (3S-5S and 3S-4D) transitions as a function of the pressure of various noble gases. The lines have a similar appearance to those of Fig. 1, with typical broadening coefficients on the order of 25 Mhz/Torr (of the same order as that observed in linear spectroscopy). A similar experiment has been carried out with He as active atoms.<sup>25</sup> In this case, absorption takes place from a metastable level. More recently, two photon absorption has been used also to study collisional processes in hydrogen states.<sup>23,24</sup>

Although two-photon absorption offers unique possibilities for high precision spectroscopic studies, the signal strengths are limited by the small excitation probability one generally encounters. The excitation probability can be enhanced if there is a nearly resonant intermediate state.

#### IV Saturated Absorption

As a more versatile alternative one can use saturation spectroscopy to obtain Doppler free line shapes.<sup>21</sup> The principle involved is quite simple and differs from that of two photon spectroscopy. In saturation spectroscopy, one uses a laser beam ("pump") to excite a given longitudinal velocity subclass of atoms and probes these atoms with a second laser beam ("probe"). Since atoms having only a narrow range of axial velocities contribute to the signal in the absence of collisions, the width of the line shape can be on the order of the natural width. The Doppler free nature of the response arises from the selection of a narrow Lorentzian velocity class, whereas in two photon spectroscopy, it is due to a cancellation of Doppler phases.

The resonance condition for saturated absorption is easily understood. The pump excites only those atoms having velocities  $kv_z = \Delta$  where  $\Delta$  is the atom-field detuning. On the other hand the probe (counterpropagating with the pump) interacts only with those atoms having  $kv_z = -\Delta$ , hence the saturated absorption signal is non-zero only for  $\Delta = 0$  (within  $\pm$  the natural width). A linear absorption background can be eliminated by suitable experimental techniques. The term "saturation spectroscopy" is used to indicate that the signal one observes is proportional to the cube of the applied field amplitude; this cubic power is composed of a quadratic term  $I^2$  representing the change of population induced by the pump field and of a factor  $I$  representing the linear absorption of the probe. One can still speak of a "weak field" limit since one is obtaining a term corresponding to the lowest nonvanishing contribution to the signal.

Narrow profiles of this type have been obtained in many atomic systems.<sup>21</sup>

In the presence of collisions the line shapes are modified.<sup>26,27</sup>

The process can be viewed as follows: the pump field excites a particular velocity sub-class of atoms, collisions cause this narrow distribution to relax towards thermal equilibrium and the probe absorption from these partially thermalized atoms gives rise to the overall line shape. Collisions enter the lineshape in two ways. First, there is the effect of collisions on optical coherence (off-diagonal density matrix elements). These effects manifest themselves as a broadening and a shift of the Lorentzian component of the absorption profile, just as in linear absorption. However, in saturated absorption there is a second effect. Collisions result in a relaxation of the non-thermal velocity distribution of the populations (diagonal density matrix elements) created by the pump field. The saturated absorption profile reflects the various collisional processes occurring in the vapor and may be used to gain information about the interatomic potential giving rise to scattering within the vapor.

In order to describe velocity changes in collisions, one generally uses a collision kernel  $W(\vec{v}' \rightarrow \vec{v})$  giving the probability density per unit time for an active atom to undergo a change from  $\vec{v}'$  to  $\vec{v}$ , as a result of collisions with the perturber bath. Although the precise form of the line shape is dependent on the specific nature of the collision kernel, general comments can be made without reference to a particular kernel. However, one must introduce the parameter  $\Delta u = \text{rms velocity change per collision}$  and  $\Gamma = \int W(\vec{v}' \rightarrow \vec{v}) d\vec{v} = \text{rate of velocity changing collisions} = \text{v.c.c.}$  The amount of thermalization is then determined by the number of collisions  $\bar{n}$  occurring within the lifetime of the transition levels and the

strength of the collision which is characterized by  $\Delta u$ . Two cases of interest occur.

1. If  $h \Delta u > \gamma$  [ $\gamma$  is the homogeneous width (NWM associated with the transition)], collisions are strong enough to remove atoms from the velocity "holes" or "bumps" created by the pump field. In this case, collisions lead to a broad background in the saturated absorption profile and the line shape can be used to monitor these v.c.c. As the perturber pressure is increased, the line shape tends towards a Voigt profile whose Gaussian component is provided by collisionally thermalized active atoms.

2. If  $h \Delta u < \gamma$ , v.c.c. are so weak that they do not significantly alter the line shape. An interesting effect can arise at low pressures. At such pressures,  $\gamma$  is determined by the natural width and one can have  $h \Delta u > \gamma$  for relatively small  $\Delta u$ . This condition leads to distorted profiles, which nevertheless may appear Lorentzian in nature. As the pressure is increased  $\gamma$  is determined by the collision broadened homogeneous width and the limit  $h \Delta u < \gamma$  is achieved. At these pressures, the line is a Lorentzian with NWM  $\gamma$ . A non-linear variation of the line width with pressure can occur for these relatively weak collisions.

All these effects can be seen in the data shown in Fig. 2 and Fig. 3. In Fig. 3, the saturated absorption profile of the 557 nm Kr I line is shown, for Kr active atoms perturbed by He.<sup>28</sup> One can identify three components in the profile: (1) A narrow Lorentzian arising from atoms not having undergone v.c.c. (2) A component arising from Kr-He collisions leading to a broad, completely thermalized Gaussian background. (3) A background arising from Kr-He collisions having  $h \Delta u > \gamma$ . In Fig. 3

is also shown the variation with pressure of the width of the components arising from v.c.c. with He and Ar atoms, respectively. One notes that both widths approach the thermal one with increasing pressure and that  $\Delta u$  for Ar perturbers is greater than  $\Delta u$  for He perturbers.

In Fig. 4, the variation with pressure of the "Lorentzian" component width  $2\gamma$  for saturated absorption in the 3.51  $\mu$ m Xe I line is shown, for Xe active atoms perturbed by Xe.<sup>29</sup> For comparison the corresponding width  $2\gamma_0$  of linear absorption is also displayed.<sup>7</sup> The saturated absorption data are strongly suggestive of the presence of the weak v.c.c. described above. Similar non-linear variations have been obtained in other systems.<sup>30-36</sup>

Experimental values for cross sections and rates of v.c.c. (representing collisions with larger impact parameters) can be orders of magnitude larger than hard sphere values.

It should be noted that saturated absorption can also be performed using two independent counterpropagating laser beams, the pump having a fixed frequency and the probe a variable one. The line shape in this case is similar to that encountered in three-level systems discussed in the next section. One can use modifications of saturated absorption experiments that have the potential of yielding higher signal-to-noise ratios. The most popular of these techniques is polarization spectroscopy.<sup>21,29</sup>

## V. Three-Level System

One slight disadvantage of saturated absorption collisional studies is that the v.e.c. effects occur in the populations of both states of the transition and it is sometimes difficult to isolate the effect on a given level. This situation can be rectified if one studies collisions using three-level systems<sup>10-12, 21</sup>, although many of these experiments require an additional laser. In a typical experiment, one starts with population in level 1 (Fig. 5), applies a field of frequency  $\Omega$ , amplitude  $\mathcal{E}$  to drive the 1-2 transition which has a frequency  $\omega$  and in addition, applies a second field of frequency  $\Omega'$ , amplitude  $\mathcal{E}'$  to complete transitions to level 3 (the 2-3 transition frequency being  $\omega'$ ). The  $\lambda_i$  represent incoherent pumping rates for the various levels and the  $\gamma_i$  decay rates from those levels. In general, the pump detuning  $\Delta = (\Omega - \omega)$  is fixed and the population of level 3 is monitored as a function of the probe detuning  $\Delta' = (\Omega' - \omega')$ . There are many variations of this three-level system (TIS) but, to be specific, we shall consider the case of the upward cascade in Fig. 5a with  $\lambda_2 = \lambda_3 = 0$ ,  $\lambda_1 \sim 0$ ,  $\gamma_1 \sim 0$ ,  $\lambda_1/\gamma_1 = \text{constant}$  to simulate the case where level 1 is the ground state. One might note, however, that high resolution line shapes can also be obtained when levels other than level 1 are incoherently pumped.<sup>6</sup>

In the absence of collisions, there is only one resonance condition for excitation to level 3 for each velocity subclass of atoms. This condition is simply a "two-photon" resonance requirement  $\Delta' = -\Delta + (h^2/c^2)v_z$  where  $c = 1$  for counterpropagating and  $c = -1$  for counterpropagating fields.

There remains some freedom in satisfying this resonance condition by the choice of the velocity subclass  $v_z$ . If  $|\Delta| \gg \hbar u$  (large detunings) one obtains maximum excitation by choosing  $v_z$  at the center of the Doppler profile ( $v_z = 0$ ). This leads to the resonance condition  $\Delta' = -\Delta$ . In this case the line shape is the convolution of a Lorentzian of width  $2\gamma_{13}$  ( $\gamma_{13} = (\gamma_1 + \gamma_3)/2$ ) and a Gaussian of width  $1.66 (\hbar + \hbar k')u$  where  $u$  is the most probably active atom velocity. One notes that if  $c = -1$  and  $\hbar \approx \hbar'$  the width of the resonance is approximately equal to the natural width  $2\gamma_{13}$  associated with the two photon transition. This width can be extremely narrow, especially if level 3 is metastable. The Doppler free nature of the line under such conditions arises from the cancellation of the Doppler phases associated with the 1-2 and 2-3 transitions.

If  $|\Delta| < \hbar u$  (small detunings) one obtains a maximum excitation by choosing those atoms having velocity  $v_z = \Delta/\hbar$  such that they are resonant with the pump field  $\Omega$ . Substituting this value of  $v_z$  into the resonance condition one obtains  $\Delta' = (\hbar k'/\hbar)\Delta$  as the position for maximum probe absorption. The line width is always on the order of the natural width of the states involved in the transitions. The Doppler free nature of this line, however, arises from the fact that only atoms having a narrow range of longitudinal velocities are contributing to the signal (there can be an additional narrowing of the line if counterpropagating waves are used, owing to Doppler phase cancellation). For either small or large detunings the probe absorption is proportional to  $\mathcal{E}^2 \mathcal{E}'^2$ . For the case of intermediate detunings it is possible to observe resonances at both

$$\Delta' = -\Delta \text{ and } \Delta' = (\hbar^2 k^2 / 2m) \Delta.$$

To include collisional effects it is more convenient to look at large and small detunings separately.

1.  $|\Delta| \gg \hbar \omega$  With collisions present, a  $\hbar \omega$  resonance centered at  $\Delta' = 0$  can result from a collisionally aided radiative excitation of level 2 by means of the reaction  $A_1 + P + \hbar \omega \rightarrow A_2 + P$  where  $A_1$  is the active atom in state 1 and  $P$  is the perturber. The difference in energy between  $\hbar \omega$  and  $\hbar \omega'$  is now compensated by a corresponding change in the atom's kinetic energy following a collision. With collisionally aided excitation of level 2, probe absorption on the 2-3 transition centered at  $\Delta' = 0$  can occur. In the absence of collisions, there is only one resonance centered at  $\Delta' = -\Delta$  and in the presence of foreign gas perturbers, a new resonance appears at  $\Delta' = 0$  which grows with increasing pressure. The width and shift of the  $\Delta' = -\Delta$  resonance can be used to obtain the 2-3 broadening and shift coefficients. Moreover, the amplitude of the  $\Delta' = 0$  resonance is proportional to the 1-2 broadening coefficient. Recent experimental data<sup>50</sup> on He ( $3^2S_{1/2} - 3^2P_{1/2} - 4^2D_{3/2}$ ) perturbed by the He are shown in Fig. 6. The effects of collisions for this large detuning case ( $\Delta = -4.0$  ku,  $\hbar/\hbar' = 1.0375$ ,  $\epsilon = -1$ ) are clearly seen (the second narrow resonance centered at  $\Delta' = 5.77$  ku arises from ground state hyperfine structure).

2.  $|\Delta| \lesssim \hbar \omega$ . The pump laser excites a given velocity subclass of level 2 population which collisions tend to thermalize. By studying the profiles as a function of pressure, it is possible to obtain in-

formation on the interatomic potential giving rise to the relaxation.<sup>51-56</sup> The following interactions occur when collisions are present

$$A_1 + P + \hbar \omega \rightarrow A_2 \quad (v_1' = \Delta/\hbar) + P$$

$$A_2 \quad (v_2' = \Delta/\hbar) + P \rightarrow A_1(v_1) + P$$

Collisions result in an excitation of level 2 and a partial thermalization of the velocity distribution. The degree of thermalization is determined by the number of collisions  $\bar{n} = \Gamma_2/\gamma_2$  ( $\Gamma_2$ : collision rate) occurring within the lifetime of level 2 and the rms change in velocity per collision  $\Delta u$ . In addition, the structure of the velocity redistribution may be used to infer something about the interatomic potential giving rise to scattering within the sample.

Theoretical probe absorption profiles for a weak pump field are shown in Fig. 7 for several pressures using the Keilson-Storer collision kernel.<sup>57</sup> One may note the gradual thermalization with increasing perturber pressure. The area under the curve remains constant. Systematic experiments of this nature were recently carried out by Bréchinac et al.<sup>58</sup> in Kr perturbed by He (Fig. 8), and by Liao et al.<sup>59</sup> in Xe perturbed by noble gases (Fig. 9). In both figures, one can see a collision-induced partial thermalization of the sample.

A complementary experiment performed using Kr and Xe was published recently.<sup>60</sup> In that system transfer from an excited state of Kr to an excited state of Xe can be achieved by quasi-resonant collisional excitation transfer. The Kr excited state is pumped in a velocity selective

## Footnotes and References

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number and the excited state is probed. Results show that the Xe atoms retain some memory of the Kr velocity, despite the fact that the Xe atoms were characterized by a Gaussian distribution before the transfer.

By careful studies of the line shapes associated with TIS, one ultimately hopes to be able to test various models for the interatomic potentials. There is some evidence from the data of Liao et al.<sup>59</sup>, for example, that suggests a hard sphere model is not sufficient to explain large angle scattering between Na ( $3P$ ) and Kr or Xe perturbations. One expects that future studies involving TIS will provide additional insight into one's understanding of collisional processes occurring in atomic systems.

## VI. Conclusion

We have briefly discussed some of the ways in which the study of laser spectroscopic line shapes associated with atomic states can be used to provide collisional data. By necessity, many interesting aspects of collisional processes and line shape formations have not been discussed at all. Among these are inelastic collisions, resonant exchange collisions, collision-induced magnetic relaxation, polarization spectroscopy, laser assisted collisional excitation, collisionally-aided radiative excitation, resonance fluorescence, strong field effects (a.c. Stark splitting), coherent transient methods, and laser spectroscopy using atomic beams. Nevertheless we hope to have conveyed a sense of the type of information one can hope to obtain from experimental line shapes of laser spectroscopy.

# References

1. I.M. Beterov and V.P. Chetochay, *Prog. Quant. Elect.* **3**, 1 (1974).
2. P.R. Berman, *Appl. Phys. (Germany)* **6**, 783 (1975).
3. K. Shimoda, *High Resolution Laser Spectroscopy*, Ed. K. Shimoda (Springer-Verlag, New York), p. 11 (1976).
4. B. Steinhilber, *J. Phys.* **3**, 19, 761 (1977).
5. P.R. Berman, *Advances in Atomic and Molecular Physics*, Eds. D.R. Bates and B. Bederson (Academic Press, New York) **23**, 57 (1977).
6. P.R. Berman, *Physics Reports* **43**, 101 (1978).
7. B. Vetter and E. Marié, *J. Phys.* **3**, 11, 2845 (1978).
8. J. Brochard and B. Vetter, *J. Phys.* **3**, 1, 315 (1974).
9. J. Brochard and B. Vetter, *J. de Physique* **38**, 121 (1977).
10. Ph. Cahuzec and R. Demachini, *J. Phys.* **3**.
11. P.R.G. Baird, K. Burnett, R. Demachini, D.M. Stacey, and R.C. Thompson, *J. Phys.* **3**, 12, 2143 (1978).
12. O. Vallee, E. Marié, N. Tran Minh, and B. Vetter, *J. Phys.* **3**.
13. R.E.M. Hodges, D.L. Drummond and A. Gallagher, *Phys. Rev. A* **6**, 1519 (1978).
14. V.P. West and A. Gallagher, *Phys. Rev. A* **11**, 1431 (1978).
15. E. Niemax, *Proc. of the 4th Int. Conf. on Spectral Line Shapes*, Ed. V.E. Baylis (Univ. of Windsor, Ontario) (1978).
16. D.E. Roberts and Z.N. Fortson, *Phys. Rev. Lett.* **31**, 1593 (1973).
17. D. Fritchard, J. Apt. and T.M. Lucas, *Phys. Rev. Lett.* **32**, 641 (1974).
18. F. Biraben, B. Cagnac and G. Grynberg, *Phys. Rev. Lett.* **32**, 643 (1974).

# References - Con't.

19. M.D. Levenson and N. Bloembergen, *Phys. Rev. Lett.* **32**, 645 (1974).
20. Th. W. Hänsch, K.C. Harvey, G. Meisel, and A.L. Schawlow, *Opt. Commun.* **11**, 50 (1974).
21. B. Cagnac, G. Grynberg and F. Biraben, *J. de Physique* **34**, 845 (1973); G. Grynberg, *Thesis*, Paris CNRS A.O. 12497 (1976).
22. For extensive references, see *Laser Spectroscopy I&II*, Ed. J.L. Hall and J.L. Carlsten (Springer Verlag, New York), (1977); *Non-linear Laser Spectroscopy*, Ed. V.S. Letokhov and V.P. Chetochay (Springer Verlag, New York) (1977); *Laser Spectroscopy of Atoms and Molecules*, Ed. H. Walther, (Springer Verlag, New York) (1976); *Frontiers in Laser Spectroscopy*, Ed. R. Balian, S. Haroche, and S. Liberman (North Holland, Amsterdam) (1977). See also Reference 3.
23. F. Biraben, B. Cagnac, E. Giacobino and G. Grynberg, *J. Phys.* **3**, 10, 2369 (1977).
24. K.H. Weber and K. Niemax, *Opt. Commun.* **18**, 317 (1979).
25. T.F. Gallagher, V.E. Cooke and S.A. Edelstein, *Phys. Rev. A* **11**, 125 (1978) and *ibid.* 904 (1978).
26. M.M. Salour, *Phys. Rev. A* **11**, 614 (1978).
27. P.W. Smith and Th. Hänsch, *Phys. Rev. Lett.* **28**, 740 (1972).
28. T. Kan and G.J. Volga, *I.E.E.E. J. Quant. Elect.* **7**, 141 (1971).
29. C. Bréchignac, B. Vetter and P.R. Berman, *Phys. Rev. A* **11**, 1609 (1978).
30. Ph. Cahuzec, E. Marié, O. Robaux, B. Vetter and P.R. Berman, *J. Phys.* **3**, 11, 645 (1978).

References - Con't.

30. G.M. Bagoev, E.V. Bakhinov, and V.P. Chebotayev, *J.E.T.P. Lett.* **16** 9 (1972).
31. T.W. Meyer, C.K. Rhodes, and H.A. Haus, *Phys. Rev. A* **12**, 1993 (1975).
32. Ph. Cahuzac, O. Robaux and R. Vetter, *J. Phys.* **B2**, 3165 (1976)
33. A.Z. Matlich, H.A. Kurnit, and A. Javan, *Chem. Phys. Lett.* **38**, 176 (1976).
34. C.J. Bords in *Laser Spectroscopy III*, Eds. J.L. Hall and J.L. Carlsten (Springer Verlag, New York) (1977).
35. I. Colomb and M. Dumont, *Opt. Commun.* **21**, 143 (1977); I. Colomb, *These du 3<sup>e</sup> cycle, Paris-Nord* (1977).
36. J.L. LeGouet, *J. Phys.* **B11**, 3001 (1978).
37. M. Bornstein and W.E. Lamb, *Phys. Rev. A* **5**, 1311 (1972).
38. S. Avrillier, Thesis, Université de Paris-Nord (1978)
39. C. Wieman and Th. W. Hänsch, *Phys. Rev. Lett.* **36**, 1170 (1976).
40. G.E. Botkin, S.F. Rautian, and A.A. Feoktistov, *Zh. Exsp. Teor. Fiz.* **52**, 1673 (1967) [*Sov. Phys. J.E.T.P.* **25**, 1112 (1967)].
41. M.S. Feld and A. Javan, *Phys. Rev.* **177**, 540 (1969).
42. Th. W. Hänsch and P.E. Toschek, *Z. Phys.* **236**, 213 (1970).
43. J.E. Bjorkholm and P.F. Liao, *Phys. Rev. A* **14**, 751 (1976).
44. D.L. Huber, *Phys. Rev.* **178**, 93 (1969).
45. A. Ouent, E.V. Smith and J. Cooper, *Astrophys. J.* **173**, 185 (1972).
46. J.L. Carlsten and A. Schöke, *Phys. Rev. Lett.* **36**, 667 (1976) and *J. Phys.* **B2**, 1231 (1976).

References - Con't.

47. J.L. Carlsten, A. Szoke, and M.G. Payne, *Phys. Rev. A* **15**, 1629 (1977).
48. D.L. Rousseau, G.D. Patterson and P.F. Williams, *Phys. Rev. Lett.* **34**, 1306 (1975).
49. R.D. Driver and J.L. Snider, *J. Phys.* **B10**, 595 (1977).
50. P.F. Liao, J.E. Bjorkholm and P.R. Berman, *Phys. Rev. A* **20**, 1489 (1979).
51. Th. W. Hänsch and P.E. Toschek, *I.E.E.E. J. Q. Elec.* **5**, 61 (1969).
52. I.M. Beterov, Y.A. Matuygin and V.P. Chebotayev, *Sov. Phys. J.E.T.P.* **37**, 756 (1973).
53. W.K. Bischel, P.J. Kelly and C.K. Rhodes, *Phys. Rev. Lett.* **34**, 300 (1975); *Phys. Rev. A* **13**, 1817 (1976), and *ibid.* **18** 29 (1976).
54. A. Kiel, A. Schabert, and P.E. Toschek, *Z. Phys.* **261**, 71 (1973).
55. W.K. Bischel, C.K. Rhodes, *Phys. Rev. A* **14**, 176 (1976).
56. Ph. Cahuzac, J.L. LeGouet, P.E. Toschek and R. Vetter, *Appl. Phys. (Germany)* **20**, 83 (1979).
57. J. Kellison and K. Storer, *Q. Appl. Math.* **10**, 243 (1952).
58. C. Brechignac, R. Vetter and P.R. Berman, *J. de Physique Lett.* **32**, 1231 (1978).
59. P.F. Liao, J.E. Bjorkholm and P.R. Berman, *Phys. Rev. A* **21**, 1927 (1980).
60. J.L. Ploqué and R. Vetter, *Phys. Rev. Lett.* **43**, 508 (1979).

# Figure Captions

1. Width  $L$  of the Lorentzian component of the  $3.51 \mu\text{m}$  line of Xe I  $\underline{v}_1$  perturber pressure obtained in linear spectroscopy. The insert shows the low pressure domain (data from Ref. 7).
2. FWHM of the Lorentzian component  $L$ , the Gaussian component  $G$ , and the total Voigt profile  $V$  of the  $3.51 \mu\text{m}$  Xe I line  $\underline{v}_1$  pressure. One notices a marked variation of the Gaussian width at low pressure; the corresponding temperatures associated with the widths are indicated on the ordinate of the  $G$   $\underline{v}_1$   $P$  curve. The variation of the Lorentzian width is nearly linear with a slope of  $(10.9 \pm 1.3) \text{ MHz/Torr}$  (data from Reference 7).
3. Saturated absorption of the  $557 \text{ nm}$  line of Kr I. The upper curve shows the experimental profile (solid line) for a perturber pressure of  $110 \text{ mTorr}$  of He and the curves 1, 2, 3 represent respectively the components of this line arising from Kr<sup>0</sup> atoms that have not experienced v.c.c., from Kr<sup>0</sup> atoms that have undergone v.c.c. with Kr, and from Kr<sup>0</sup> atoms that have undergone v.c.c. with He atoms. The lower graph represents the width of the v.c.c. component "3" perturbed by He perturbers (dots) or Ar ones (crosses). The dashed line is the thermal equilibrium value (data from Reference 28).
4. Width of the  $3.51 \mu\text{m}$  line of Xe I as a function of Xe pressure in both linear ( $2\gamma$ ) and saturation ( $2\gamma$ ) spectroscopy. Point "A" is the natural line width of  $4.6 \text{ MHz}$ . One observes a marked non-linear variation of the width, owing to v.c.c. (data from Reference 29).

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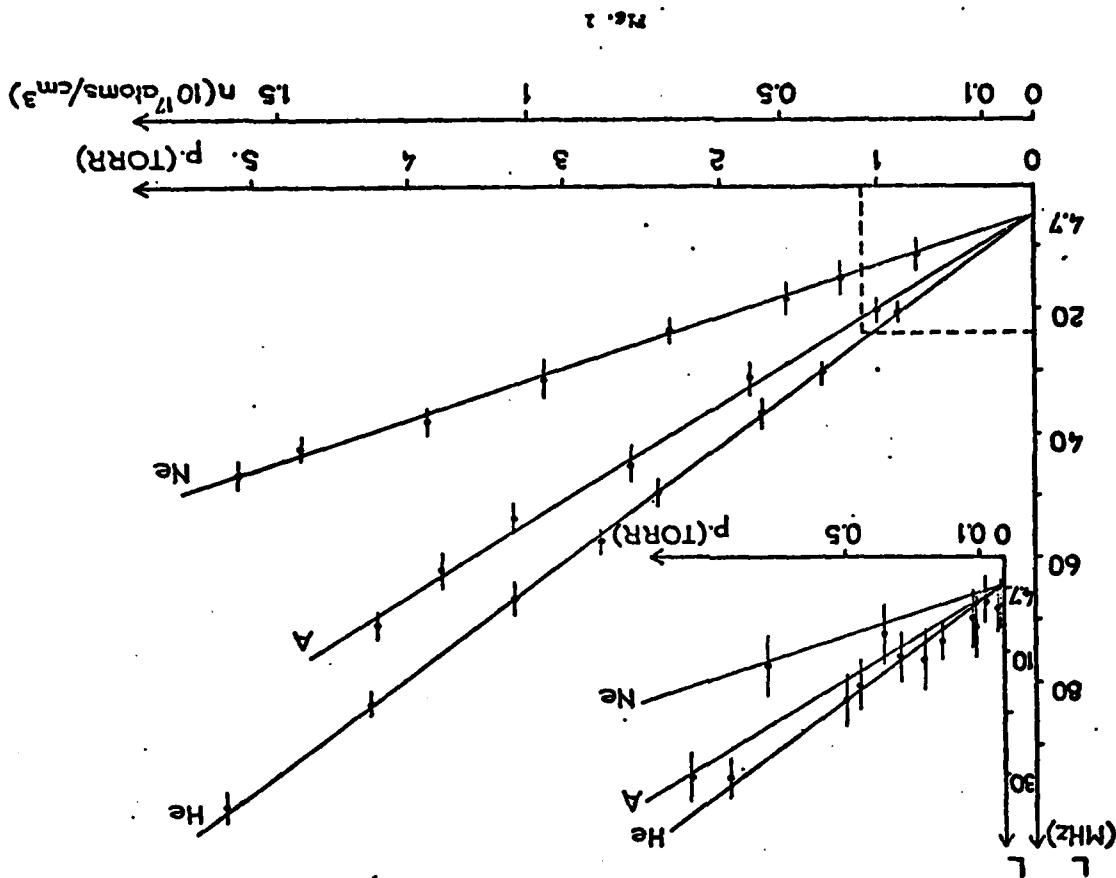
5. Three-level systems used in saturation spectroscopy. (a) upward cascade, (b) inverted "v", (c) "v". The  $\lambda_i$  are values for incoherent pumping and  $\gamma_i$  the decay rates for the states  $i$ . In this work, we consider case (a) in the limit  $\lambda_2 = \lambda_3 = 0$ ,  $\lambda_1 = 0$ ,  $\gamma_1 \neq 0$  and  $\lambda/\gamma = \text{constant}$ , simulating a system in which level 1 is the ground state.
6. Experimental profiles of probe absorption for the excitation of the  $3^2S_{1/2} - 3^2P_{1/2} - 4^2D_{3/2}$  transition in He for various pressures of He perturbers. The pump detuning is  $\Delta/2\pi = -4 \text{ GHz} = -4 \text{ km/2}\pi$ . The two narrow resonances represent "direct" two photon excitation of the  $4^2D$  state from the two hyperfine components of the ground state. The collision induced broad resonance is centered at the  $3^2P_{1/2} - 4^2D_{3/2}$  transition frequency. Solid lines, experiment; points, theoretical.. fit (data from Reference 50).
7. Theoretical profiles for probe absorption  $I_p$  (in arbitrary units) in a TLS with  $k'/k = 0.4$ , counterpropagating waves ( $c = -1$ ), a detuning  $\Delta/kv = -1$ , and a weak pump field. The population inversion  $N_{32}$  between levels 3 and 2 in the absence of collisions is taken to be zero. The abscissa is the probe detuning  $\Delta'$  in units of  $kv$ . The curves are calculated for different perturbers using a Keilson-Storer collision kernel with  $\Delta v = 0.66u$  and a v.c.c. rate  $\Gamma_2/kv = 0.006 P$ , where  $P$  is the perturber pressure in Torr. The lifetime of the intermediate state is  $\gamma_2/kv = 0.02$ . These curves show the thermalization of

Figure Captions - Con't.

the intermediate state population velocity distribution with increasing  $P$ .

8. Profiles for linear and saturation spectroscopy of the 557 nm line of KrI. Curve I, linear absorption profile in pure Kr, centered at  $\nu_0$ . Curves II and III represent saturated absorption profiles obtained with two counterpropagating laser beams, the pump being detuned by  $\Delta/\nu_0 = 600$  MHz from  $\nu_0$ . Curve II is recorded for pure Kr at a pressure of 10 mTorr and curve III is recorded for a mixture of Kr (10 mTorr) and He (450 mTorr). Curve III is analyzed in terms of components arising from Kr atoms which have not experienced v.e.c. (curve (a)), Kr atoms that have undergone thermalizing events with Kr atoms (curve (b)) and Kr atoms that have undergone v.e.c. with He atoms (curve (d)). The lower graph shows the contribution to curve (c) from Kr atoms that have undergone 1, 3, 5, ... collisions with He, indicating the gradual thermalization of Kr as a function of the number of Kr - He collisions (data from Reference 58).

9. Experimental probe absorption as a function of  $\Delta/\nu_0$  for the same transition in He shown in Fig. (6), but for a pump detuning  $\Delta/\nu_0 = -1.6$  kHz/2 $\pi = -1.6$  GHz. The partial thermalization resulting from v.e.c. is evident. The partial circles represent a theoretical fit using a hard sphere collision kernel and the open circles a fit using the Kelso-Storer collision kernel (data from Reference 59).



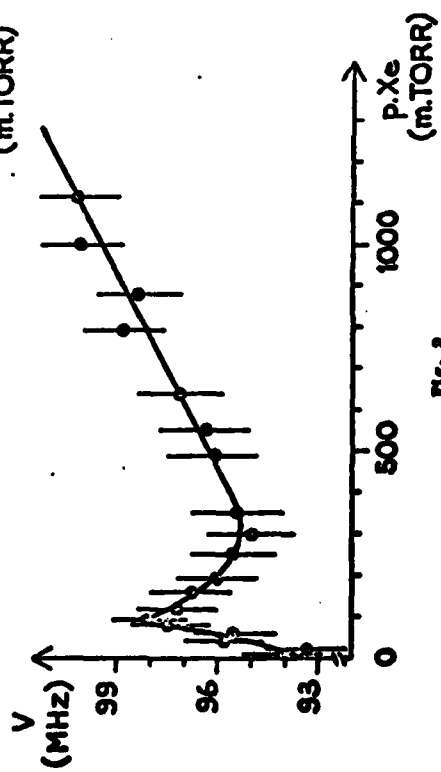
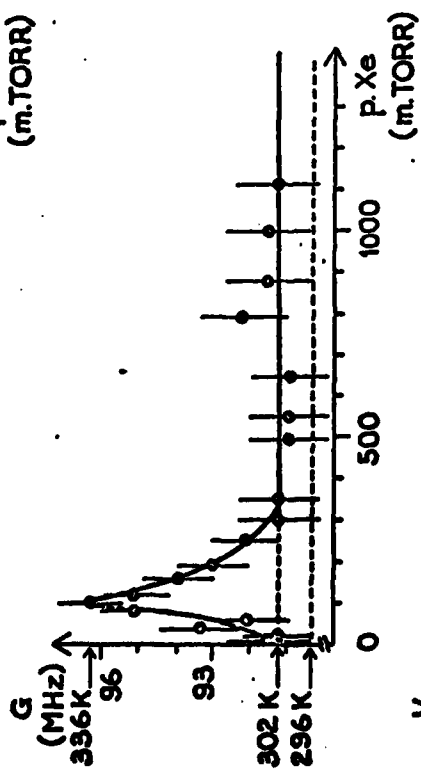
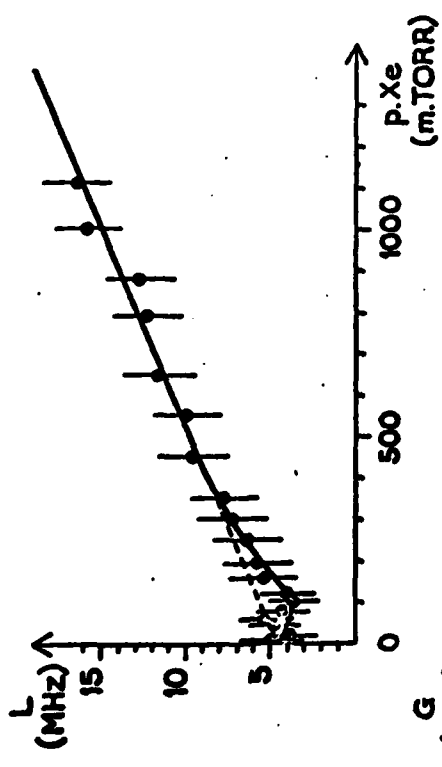


Fig. 2

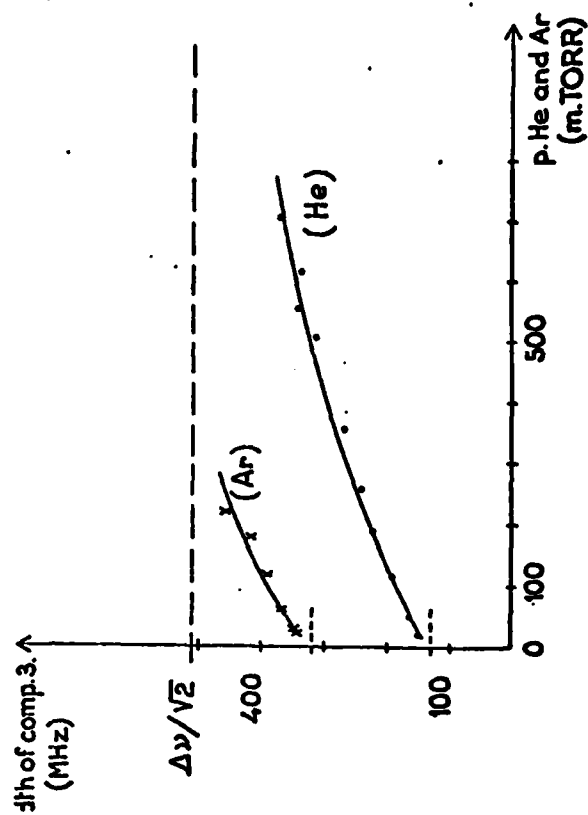
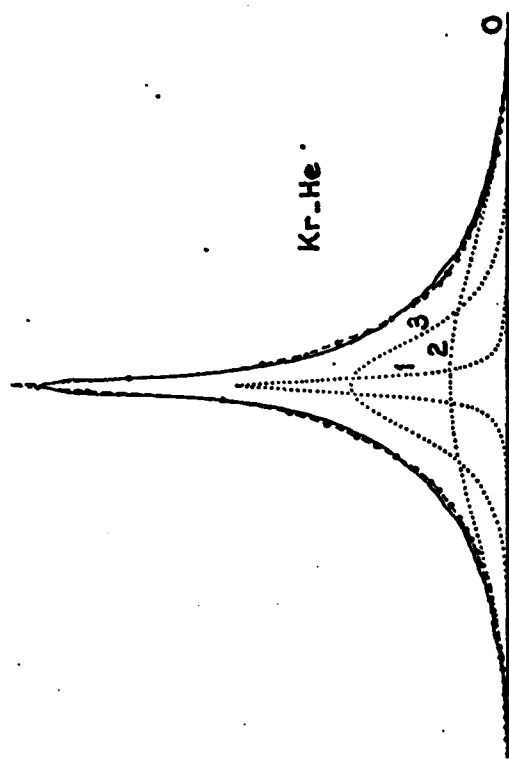


Fig. 3

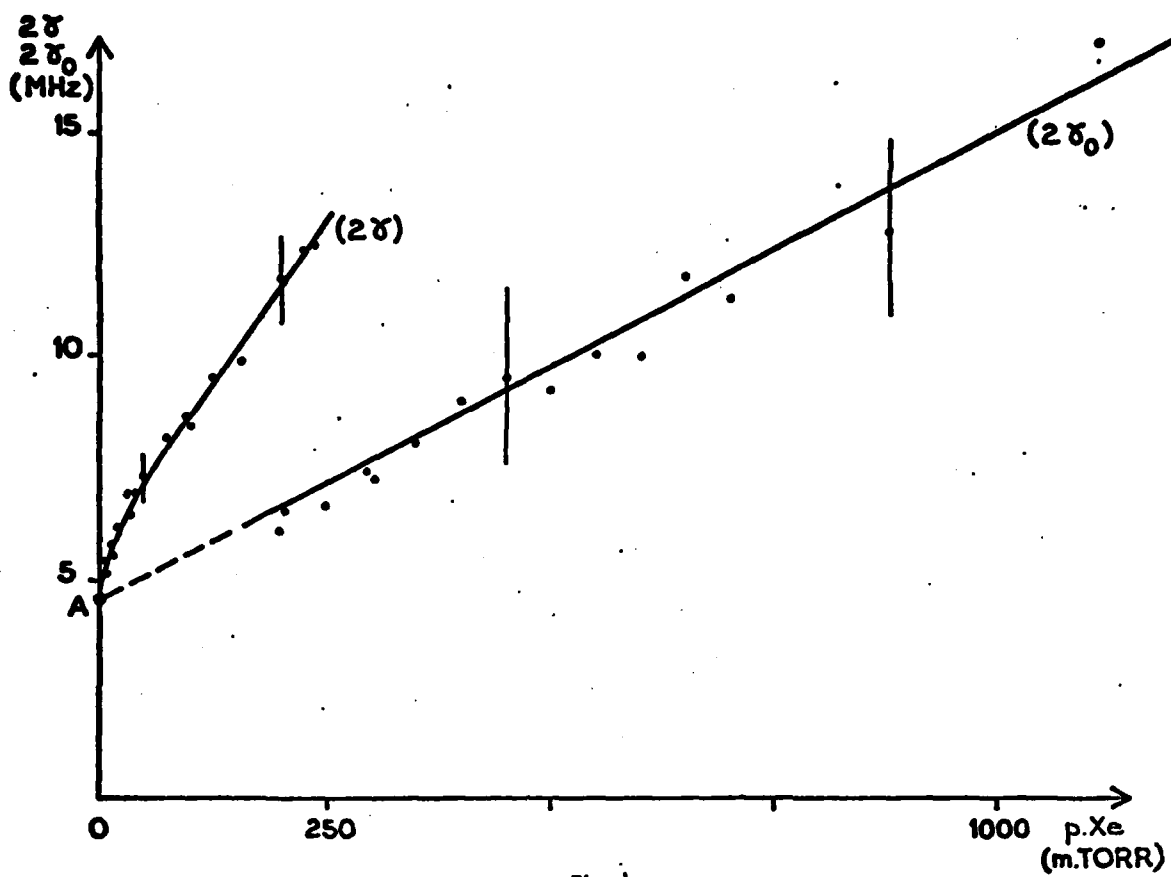


Fig. 4

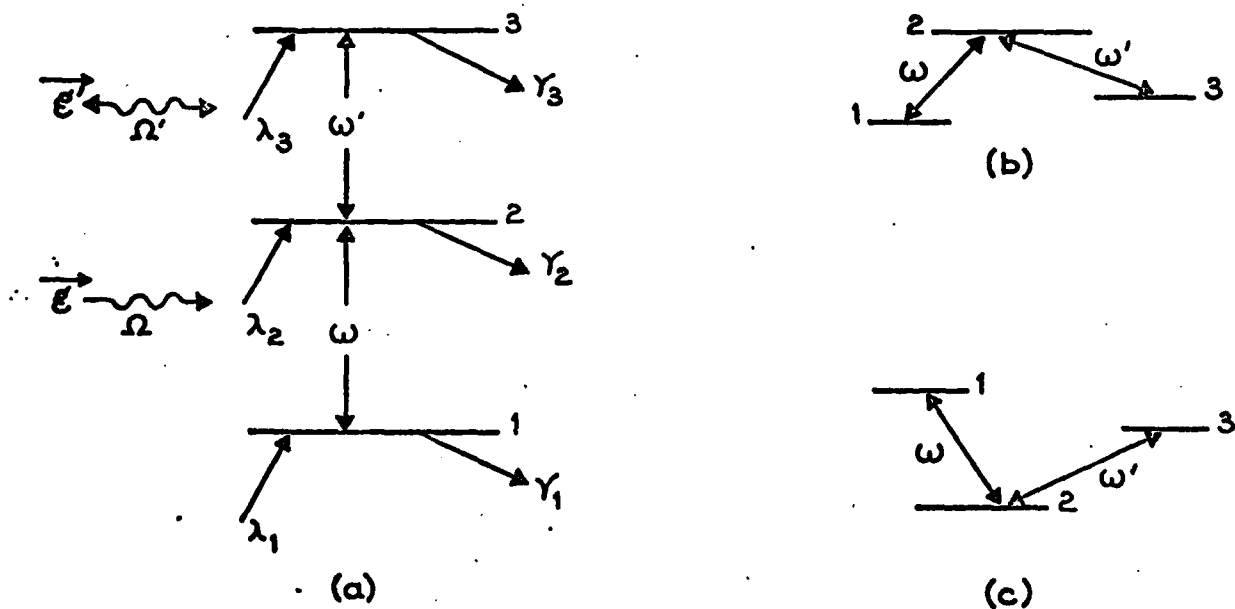


Fig. 5

$\Delta = -1$   $\epsilon = -1$

$N_{32} = 0$

$\uparrow I_3$

2.7  $p = 20$

7.3

$p = 5$

20

$p = 2$

116

$p = 0$

-2 -1 0 .4 1 2  $\Delta'$

Fig. 7

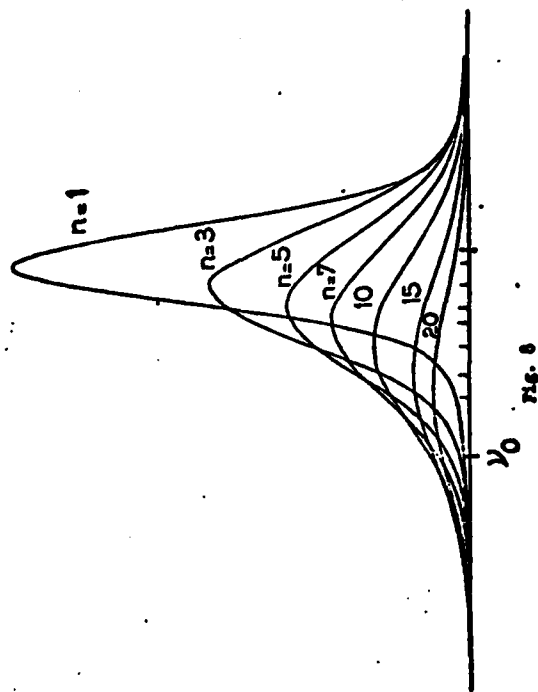
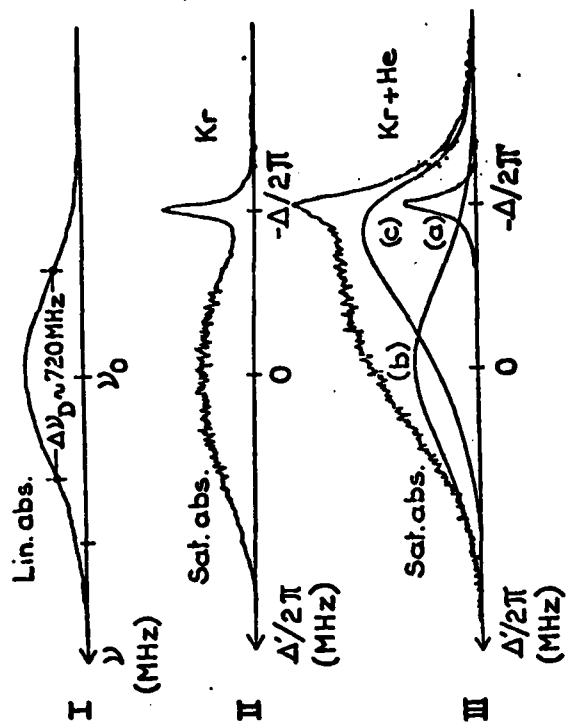


Fig. 8

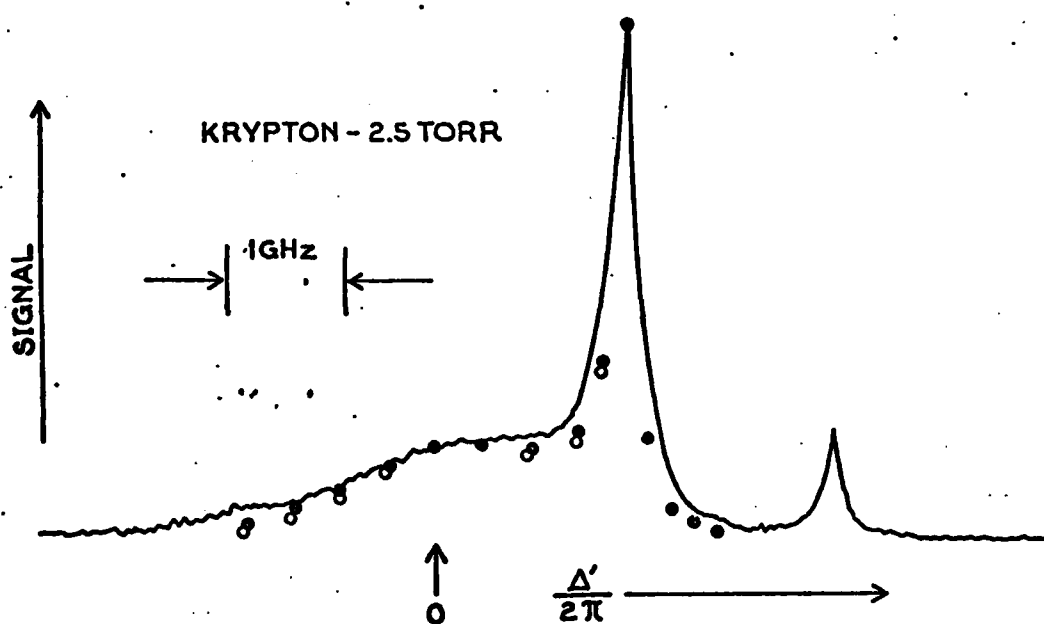


Fig. 9